Supporting Information

Ionic liquid gating and phase transition induced semiconducting to metallic transition in $La_{0.67}Sr_{0.33}MnO_3/BaTiO_3$ heterostructures

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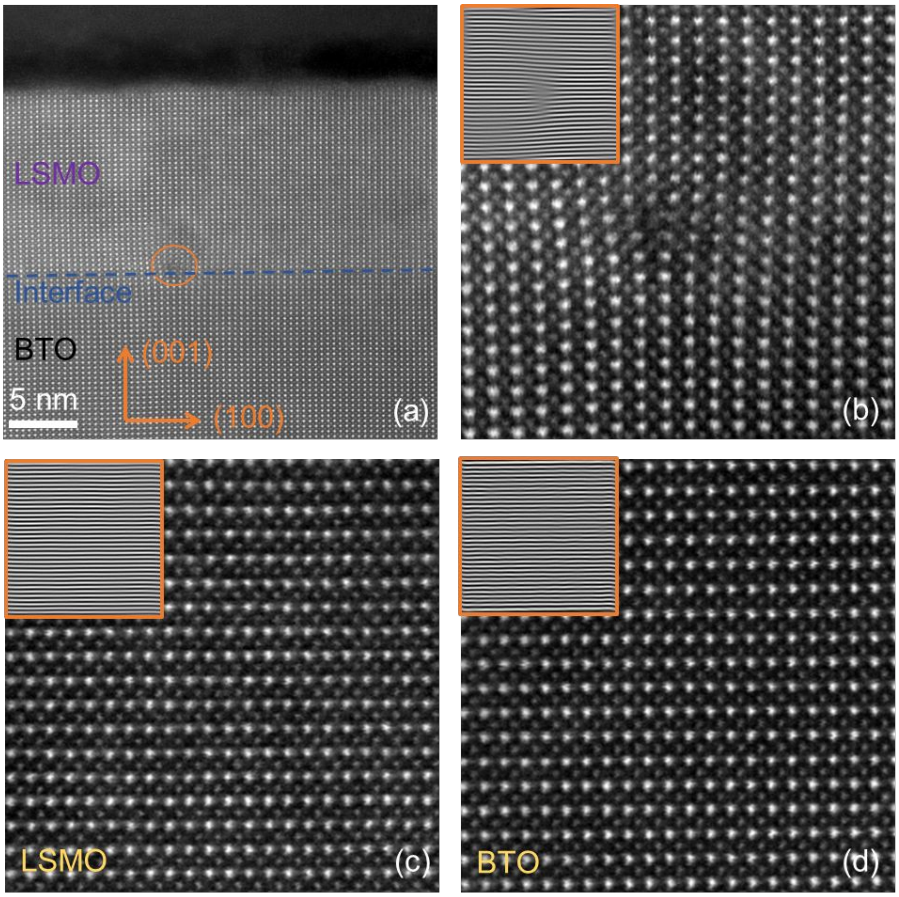

Figure S1. (a) The STEM image of LSMO/BTO heterostructure. (b), (c) and (d) are enlarged views of the interface, the LSMO film, and the BTO substrate, respectively. The corresponding inverse fast Fourier diffractograms are shown in the upper left corner inset. The defects due to large lattice mismatch can be observed at the interface.

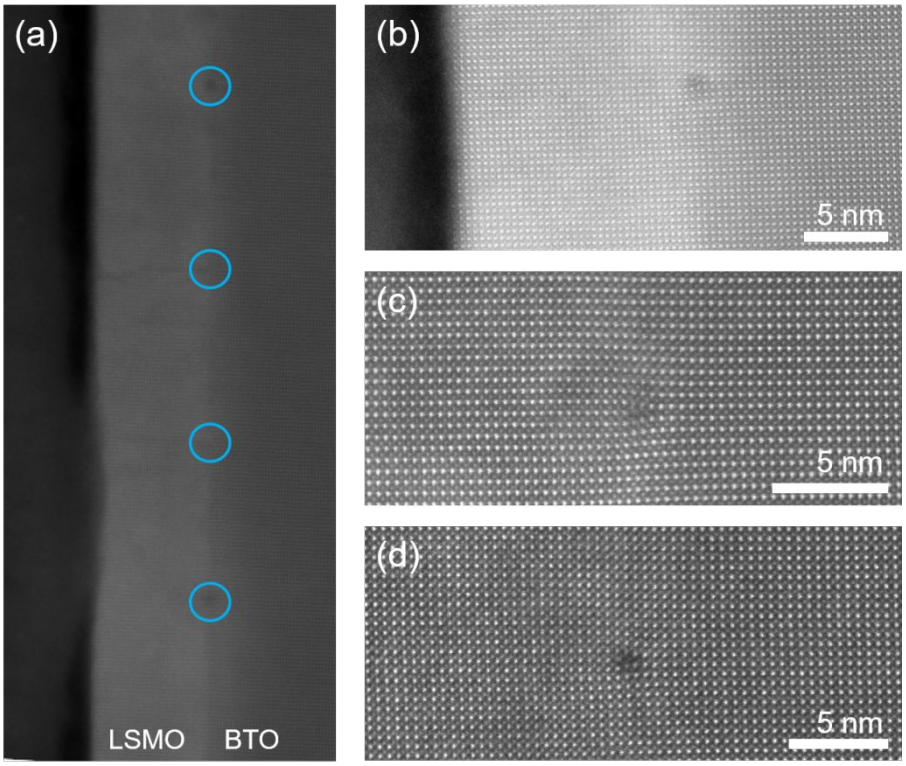

Figure S2. The STEM images of the LSMO/BTO structure. (a) The interface of the structure. The defects are highlighted by blue circles. (b), (c) and (d) is the enlarged view of the defects.

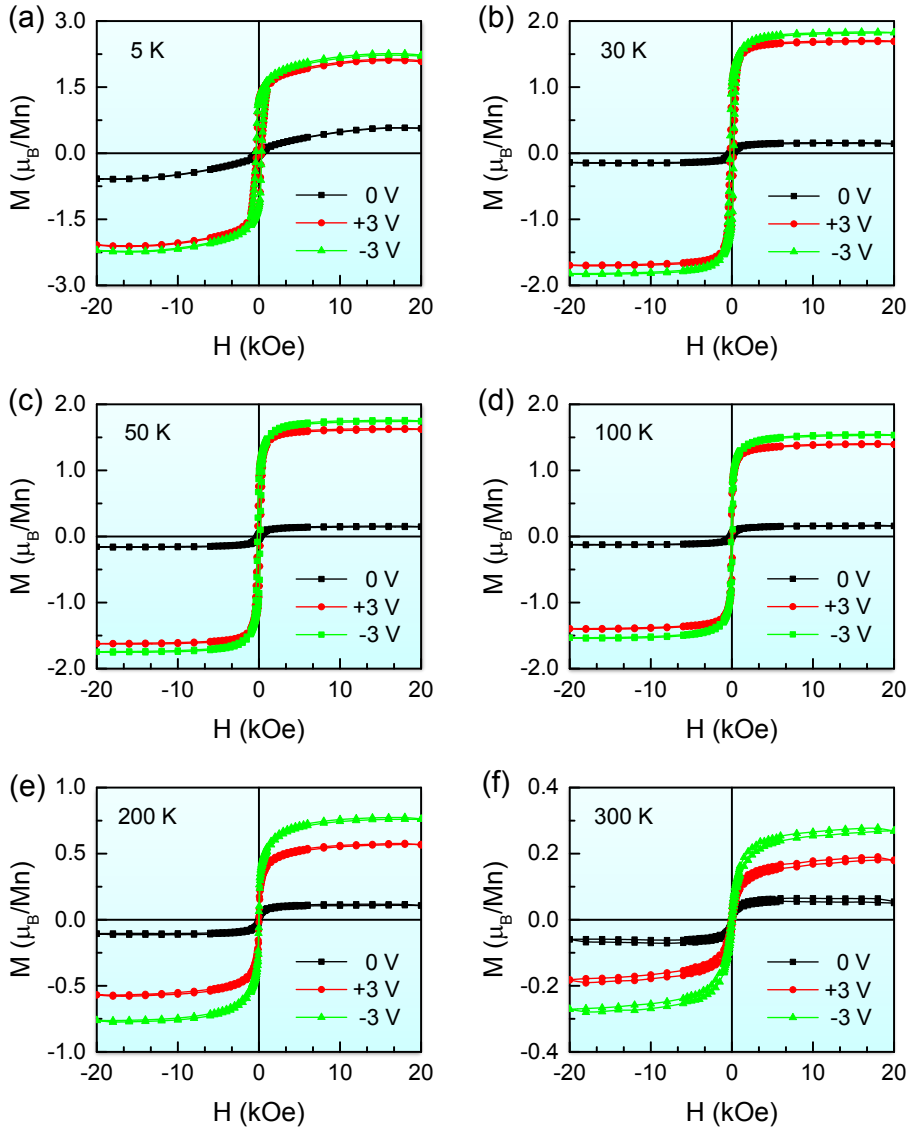

Figure S3. The *M-H* curves for LSMO/BTO heterostructures at V_G =0 V, +3 V, and –3 V, respectively, which was measured at (a) 5 K, (b) 30 K, (c) 50 K, (d) 100 K, (e) 200 K and (f) 300 K. The LSMO film at V_G =0 V shows extremely weak magnetic signals. And the films at V_G = +3 V and –3 V exhibit substantially equivalent magnetic properties at low temperatures. With the temperature increasing, the *M-H* curves at V_G = +3 V and –3 V gradually show a difference, overall, the magnetic properties of LSMO film at V_G = –3 V are relatively better.

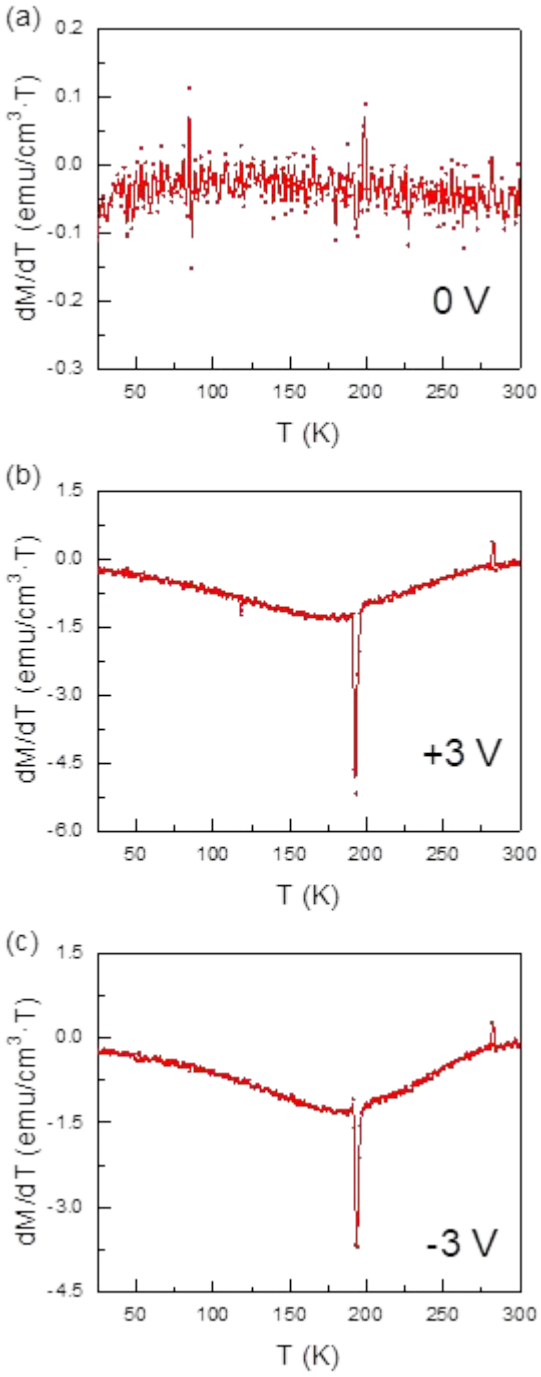

Figure S4. The derivate dM/dT, as a function of temperature under FC for LSMO/BTO heterostructures at V_G = (a) 0 V, (b) +3 V and (c) -3 V. The curve at V_G =0 V is messy, which indicates the pristine state sample is almost non-magnetic. The obvious abrupt changes for the two curves at V_G = +3 V and -3 V correspond to the phase transition temperature of the BTO substrate. Even ignoring the abrupt changes

caused by the phase transition, we find that the curve at V_G = -3 V still reaches the minimum value near the *O*→*R* phase transition temperature.

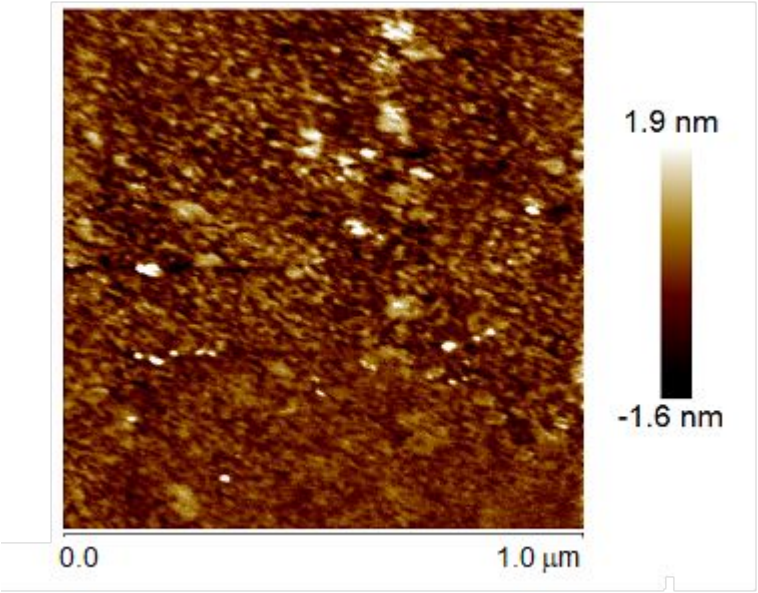

Figure S5. (a) AFM image of the gated LSMO/BTO heterostructure. Comparing with the pristine state, it becomes slightly rough. However, the surface morphology does not appear obvious damage, which indicates that the sample still has a good structure.

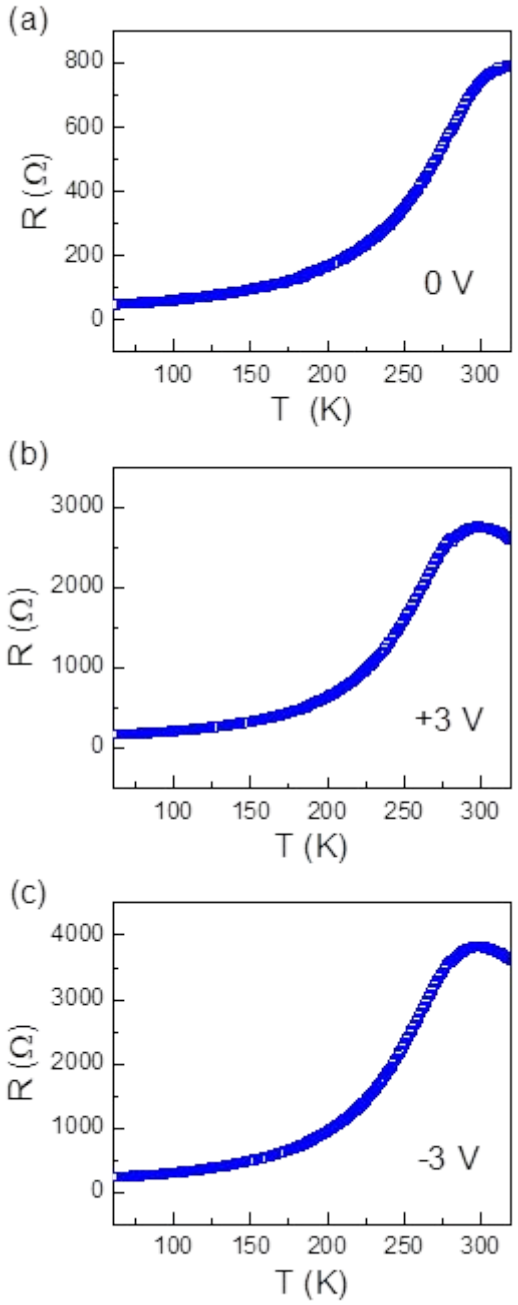

Figure S6. The *R*-*T* curves for LSMO/$SrTiO_3$ heterostructures at V_G = (a) 0 V, (b) +3 V and (c) –3 V. The measurement temperature ranges from 320 K to 60 K. All the heterostructures for three states exhibit metal-insulator transition, and the transition temperatures change with V_G. However, we can find that the regulation of positive and negative V_G on the metal-insulator transition temperature of LSMO is not obvious.

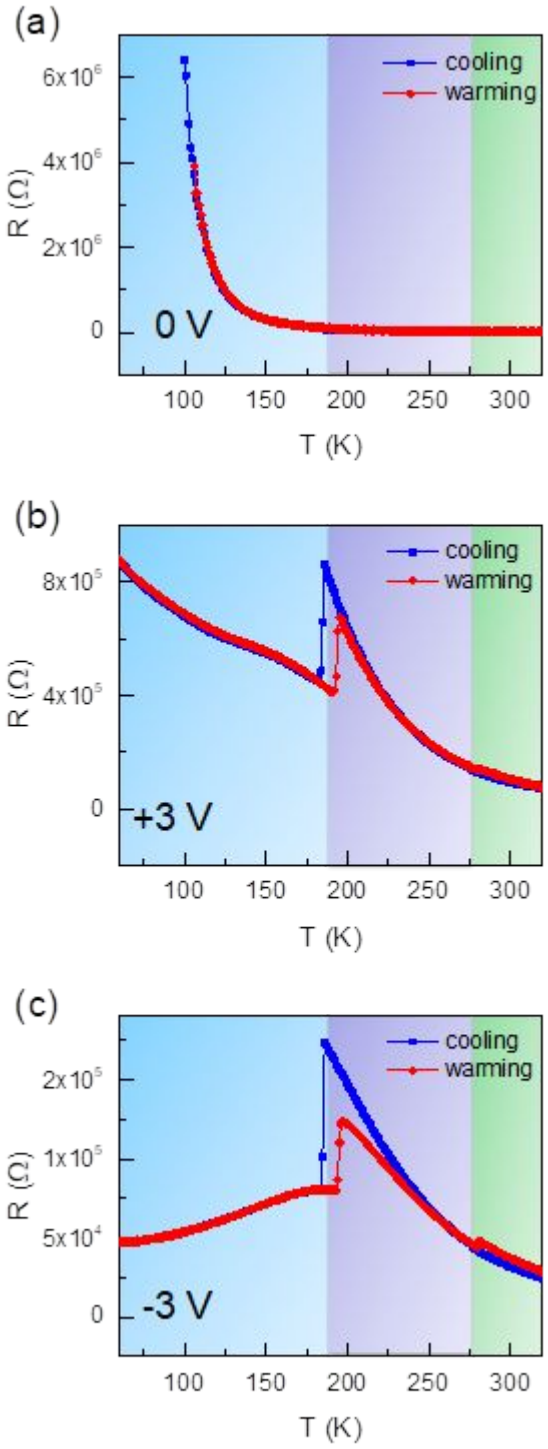

Figure S7. The *R*-*T* curves for LSMO/BTO heterostructures at V_G = (a) 0 V, (b) +3 V, and (c) –3 V, which records the data during the cooling (warming) process from 320 K to 60 K (60 K to 320 K). The three-color regions in the figure represent the temperature distribution of the three-phase regions of the BTO.

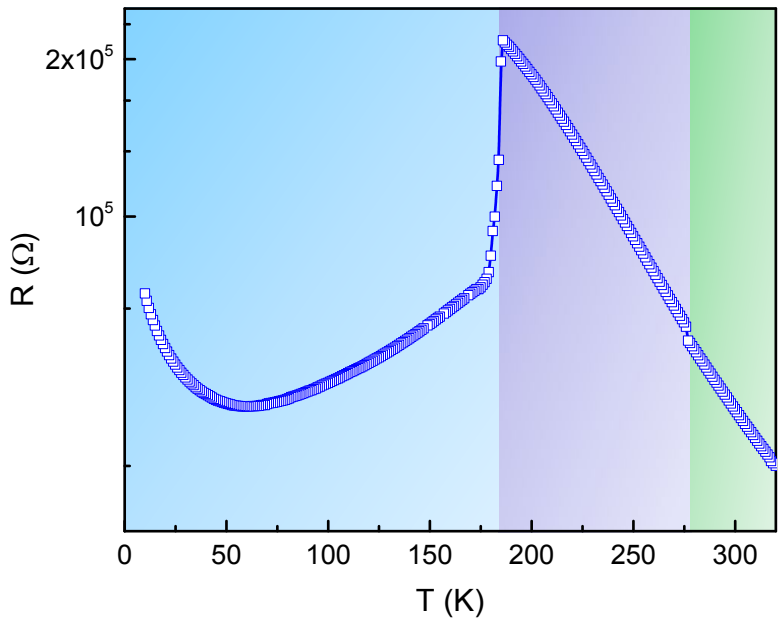

Figure S8. The *R*-*T* curves for LSMO/BTO heterostructures at V_G = –3 V, which records the data during the cooling process from 320 K to 10 K. The three-color regions in the figure represent the temperature distribution of the three-phase regions of the BTO. We can find that the resistance of LSMO film has a decreasing trend with the temperature decreases from 185 K to 60 K. As the temperature decreases further below 60 K, the resistance shows an upward trend with the decreasing temperature. This may due to the weak localization effect.

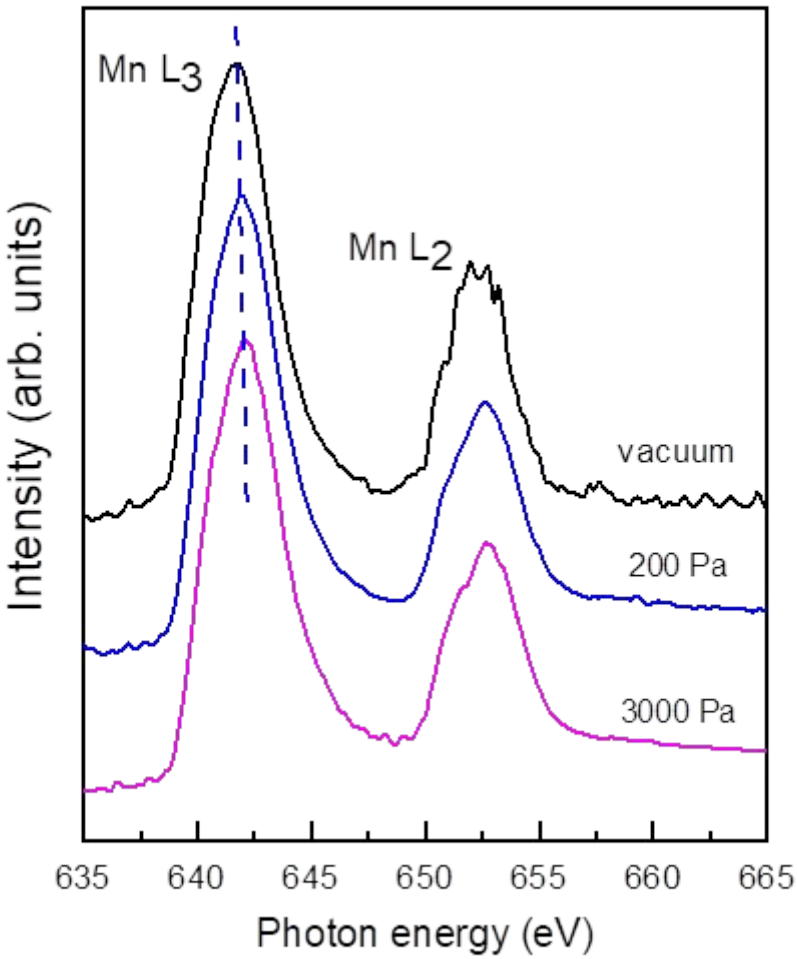

Figure S9. The XAS at Mn $L_{3,2}$ edges for 5 nm LSMO films, the films adopt three annealing forms, namely vacuum environment annealing (black line), oxygen atmosphere with 200 Pa annealing (blue line) and 3000 Pa annealing (magenta line). The black line marks the corresponding peak of the L_3 edge. As the oxygen pressure increases during annealing, the L_3 peak shift toward higher energy levels, suggesting that the valence state of Mn ions in the corresponding LSMO films increases.